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Catalytic Combustion of Residual Fuels

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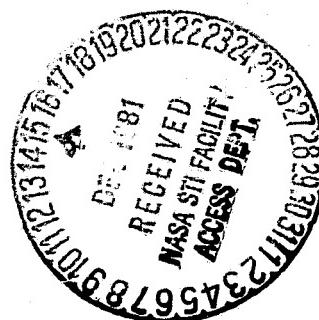
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U.S. DEPARTMENT OF ENERGY
Fossil Energy
Office of Coal Utilization



Prepared for
Fifth Workshop on Catalytic Combustion
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CATALYTIC COMBUSTION OF RESIDUAL FUELS

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ABSTRACT

A noble metal catalytic reactor was tested using two residual fuels at inlet air temperatures from 700 to 960 K, pressures of 3×10^5 Pa and 6×10^5 Pa, and reference velocities of 10, 15, and 20 m/s. Combustion efficiencies greater than 99.5 percent were obtained for both residual fuels. Steady state operation of the catalytic reactor required inlet air temperatures of at least 800 K for both residual fuels. At lower inlet air temperatures, upstream burning in the premixing zone occurred which was probably caused by fuel deposition and accumulation on the premixing zone walls. Increasing the inlet air temperature to 800 K prevented this occurrence. Both residual fuels contained about 0.5 percent nitrogen by weight. NO_x emissions ranged from 50 to 110 ppm by volume at 15 percent excess O₂. Conversion of fuel-bound nitrogen to NO_x ranged from 25 to 50 percent.

E-1040

INTRODUCTION

Lewis Research Center is currently evaluating catalytic combustion as part of the Critical Research and Advanced Technology Support Project sponsored by the D.O.E. Office of Fossil Energy, Division of Coal Utilization. Catalytic combustion has shown the potential for extremely low NO_x emissions and high combustion efficiency when operated with light distillate and gaseous fuels (Refs. 1 and 2). Catalytic combustion systems are able to complete the combustion reactions at relatively low flame temperatures compared to conventional combustors. This results in negligible thermal NO_x emission levels from fuels which do not contain significant amounts of nitrogen. Catalytic combustion has also been successfully demonstrated with coal-derived liquids (Refs. 3 and 4). These fuels typically contain reduced levels of hydrogen and increased levels of nitrogen when compared to petroleum-derived fuels.

The present study reports an experimental evaluation of catalytic combustion with two grades of petroleum-derived residual fuel. Residual fuels are typically very viscous, with high boiling points, and can also contain relatively high levels of fuel-bound nitrogen. Their use may become important for stationary gas turbine applications due to the unavailability of higher grade fuel or economic considerations. Due to the physical properties of the fuel, significant problems were anticipated in premixing and partially prevaporizing the fuel to achieve a uniform fuel-air distribution before entering a catalytic reactor. Tests at atmospheric pressure of a No. 6 fuel oil indicated problems with flashback into the premixing zone and unsteady operation of the catalytic reactor (Ref. 5). Catalytic combustion of residual fuel is also reported in Refs. 6 and 7. Reference 6 reported upstream burning in the premixing zone which made steady operation of the reactor possible only with diesel No. 2 and residual fuel oil blends. Reference 7 reported stable operation with residual fuel at inlet temperatures above 725 K.

In the present study, a noble metal catalytic reactor was tested at inlet air temperatures ranging from 700 to 960 K, reference velocities from 10 to 20 m/s, and pressures of 3×10^5 and 6×10^5 Pa. Two residual fuels were used in the study. Temperatures, catalytic reactor pressure drop and emissions of CO₂, CO, NO_x, and unburned hydrocarbons were measured.

EXPERIMENTAL DETAILS

Test Rig

A schematic drawing of the test rig used for this study is shown in Fig. 1. It was fabricated from 15.2 cm (6 in. nominal, schedule 40) diameter stainless steel pipe. Carborundum T30R fiberfrax tube insulation was inserted inside the pipe to minimize heat losses. A 0.16 cm thick stainless steel liner was inserted inside the insulation upstream and downstream of the catalytic reactor to prevent erosion of the insulation.

The inlet air was indirectly preheated and its temperature was measured at a plane just upstream of the fuel injector with an array of 12 Chromel-Alumel thermocouples mounted in a flange. Test section inlet pressure was measured at a tap located in the flange containing the inlet thermocouples. Pressure was controlled by a back pressure valve. The airflow entering the test rig was measured with a standard ASME orifice.

The fuel injector used was of a design developed by Tacina (Ref. 8). It was the hexagonal tube airblast fuel injector previously used for testing coal-derived liquids (Ref. 3). It was constructed from 19 hexagonal tube modules welded together. Figure 2(a) is a schematic drawing of one of the modules which comprise the complete fuel injector. Fuel was injected through a 0.07 cm inside diameter tube pointing downstream in the center of the smallest cross-sectional area of each module to provide good atomization and mixing. All fuel tubes were the same length, 25.4 cm, to provide equal flowrates to each module. As a precaution, to prevent plugging of the fuel tubes at the high inlet temperatures used for this study, cooling air was introduced into an outside tube which surrounded each fuel tube. All testing was performed with the cooling air, which comprised less than 5 percent of the total air flowrate. Figure 2(b) shows the inlet face of the fuel injector.

The premixing zone length of 30.6 cm was held constant for all tests. A Pt/Pt-13 percent Rh thermocouple was used to detect upstream burning in the premixing zone.

The catalytic reactor used for this study consisted of 8 elements. Each element was 12 cm in diameter and 2.54 cm long. The elements were separated by a 0.31 cm gap containing at least one thermocouple except for the last two elements which were not separated. This arrangement is shown in Fig. 1. The elements which comprise the reactor are described in Table 1. The first two elements had the largest cells, the next two smaller, with the last two having the smallest cells. This graded cell arrangement was successful in preventing plugging of the first element passages when used in previous studies with coal-derived liquids. The static pressure drop across the catalytic reactor was measured with a differential pressure transducer connected between a tap at the premixing zone thermocouple station and one located in the flange at the first row of thermocouples downstream of the catalytic reactor.

At a distance 17.2 cm downstream of the catalytic reactor, a fixed location single point water-cooled gas sampling probe, with a 0.6 cm inside diameter sampling passage, was used to withdraw samples for emissions measurements. Temperatures were also measured downstream of the reactor at the locations shown in Fig. 1. The gas sample line was electrically heated to prevent unburned hydrocarbons from condensing. Concentrations of CO and CO₂ were measured with monodispersive infrared analyses, unburned hydrocarbons with a flame ionization detector, and nitrogen oxides (total NO + NO₂) with a chemiluminescent analyzer.

The residual fuel tank and lines were electrically heated to 400 K to reduce the fuel viscosity for pumping. Normal operating procedure for testing with residual fuel was to initially start the reactor with diesel No. 2 and then gradually introduce residual fuel flow while slowly decreasing the diesel fuel flow until the reactor was operating solely on residual fuel. The procedure generally required about 10 minutes to complete. This prevented residual fuel from impinging upon relatively cold catalytic reactor walls which would tend to plug the catalyst channels. Shutdown was accomplished by reversing the above procedure to purge the residual fuel from the small fuel injector tubes.

MEASUREMENTS AND COMPUTATIONS

Reference Velocity

The reference velocity was computed from the measured mass flow rate, the average inlet air temperature, the duct cross-sectional area, and the test-section inlet pressure.

Emission Index

Emissions were measured as concentrations in ppm by volume, corrected for water of combustion, and converted to emission indices using the expressions in Ref. 9.

Combustion Efficiency

Combustion efficiency was calculated from the following expression.

$$\text{Eff} = 100 - 0.1 (\text{E.I.}_{\text{HC}} - \text{E.I.}_{\text{HC, EQ}}) - 0.1 \frac{\text{HV}_{\text{CO}}}{\text{HV}_{\text{FUEL}}} (\text{E.I.}_{\text{CO}} - \text{E.I.}_{\text{CO, EQ}})$$

where

Eff combustion efficiency, percent
E.I._x emission index of specie x, g_x/kg fuel
HV_x lower heating value of x, J/kg

Equilibrium concentrations (E.I._x, EQ) were obtained from the computer program of Ref. 10.

Fuel-Air Ratio

The fuel-air ratio was determined both from the metered fuel flow and airflow rates and by making a carbon balance from the measured concentrations of CO, CO₂, and unburned hydrocarbons. The two values agreed within ±12 percent. The adiabatic reaction temperature was computed from the computer program of Ref. 10 using the carbon balance fuel-air ratio. The carbon balance fuel-air ratio was selected since it was the local fuel-air ratio at which the emissions data were obtained. It also included the small amount (less than 5 percent of the total airflow) of air used for cooling the fuel tubes.

NO_x Emissions

Measured NO_x emissions were corrected to 15 percent O₂ using the following expression:

$$\frac{(\text{NO}_x)_{\text{15 percent O}_2}}{(\text{NO}_x)_{\text{15 percent O}_2, \text{ ppmv}}} = \frac{(\text{F/A})_{\text{15 percent O}_2}}{(\text{F/A})_{\text{C-Bal}}} (\text{NO}_x)_{\text{measured, ppmv}}$$

where

(F/A)_{15 percent O₂} is the fuel-air ratio which will produce an exhaust of 15 percent O₂.

(F/A)_{C-Bal} is the carbon balance fuel-air ratio.

RESULTS AND DISCUSSION

Two residual fuels, batch 1 and batch 2, were used for this study. Fuel properties are listed in Table II. Both fuels contained about 0.5 percent by weight of fuel-bound nitrogen. Carbon and hydrogen contents of both fuels were also very similar. The viscosity of batch 2 residual fuel was considerably greater than batch 1. At 340 K, the viscosity of batch 2 was about 4 times greater than batch 1. Complete distillation curves were not obtained for the residual fuels. For batch 1 residual fuel, 15 percent of the fuel was distilled at 608 K. For batch 2 residual fuel, no distillation was obtained for temperatures up to 589 K.

Combustion Efficiency

Combustion efficiency, as a function of adiabatic reaction temperature, is presented in Figs. 3(a) and (b). Data are presented in Fig. 3(a) for both residual fuels at a reference velocity of 10 m/s for pressures of 3x10⁵ and 6x10⁵ Pa, and inlet air temperatures of 800, 900, and 960 K. Combustion efficiencies greater than 99.5 percent were obtained for adiabatic reaction temperatures greater than 1350 K. Batch 1 residual fuel had a greater combustion efficiency than batch 2 for a given adiabatic reaction

temperature. This was probably caused by a lower percentage of batch 2 fuel being vaporized. Reference 6 reported an effect of fuel vaporization on combustion efficiency. There are two reasons for batch 2 fuel having a lower percentage of fuel vaporized than batch 1. First, the greater viscosity of batch 2 would result in larger drop sizes (Ref. 11) and thus a lower vaporization rate. Second, less vaporization at a given temperature would occur for batch 2 residual fuel because of its increased distillation temperature.

Figure 3(a) also shows an effect of pressure on combustion efficiency. For batch 2 residual fuel, combustion efficiency was greater at a pressure of 6×10^5 Pa than at 3×10^5 Pa at adiabatic reaction temperatures less than 1400 K. Increasing pressure and inlet temperature increase gas phase reactions (Refs. 12 and 13). Increasing inlet temperature also initiates gas phase reactions sooner in the catalytic bed (Ref. 13). For batch 2 residual fuel, preflame reactions in the premixing zone were increased at the higher pressure. At a reference velocity of 10 m/s, the premixing zone temperature was 1050 to 1100 K at a pressure of 6×10^5 Pa and 950 to 1000 K at a pressure of 3×10^5 Pa, independent of the inlet air temperature. Since this is the inlet gas temperature to the catalytic reactor, the increased inlet temperature would initiate gas phase reactions sooner in the catalytic bed which would then proceed at a faster rate because of the increased temperature and pressure.

At a constant pressure, little effect of inlet air temperature is shown. Reference 13 reported an increase in combustion efficiency for increased inlet air temperatures. Preflame reactions in the premixing zone were observed with the residual fuel which resulted in a relatively constant fuel-air mixture temperature at the catalytic reactor inlet at a constant pressure. This could have eliminated the expected increase in performance with increased inlet air temperature.

Figure 3(b) presents combustion efficiency data at a reference velocity of 20 m/s and a pressure of 3×10^5 Pa for both residual fuels. Increasing reference velocity decreased combustion efficiency. At an inlet air temperature of 800 K, an increase in fuel-air ratio corresponding to an adiabatic reaction temperature increase of about 50 K is required over that needed at 10 m/s to maintain the same combustion efficiency.

No data is presented for inlet air temperatures below 800 K. Unstable operation of the system resulted for inlet air temperatures below 800 K. For batch 1 residual fuel, at an inlet air temperature of 750 K, the premixing zone thermocouple would rise from slightly less than 750 K to about 1400 K at intervals of approximately 8 minutes. A sharp increase in CO₂ and decrease in CO emissions would result from the temperature excursion. Between these perturbations, catalytic reactor performance was quite stable. At an inlet air temperature of 700 K, operation became more unstable with the premixing zone thermocouple rising from about 700 to 1500 or 1600 K at intervals of approximately 1 or 2 minutes. Emissions would also fluctuate at about the same rate. It seems likely that fuel was depositing on the premixing zone duct walls at the reduced inlet air temperature, accumulating, and then igniting. Inlet air temperatures of 800 K and above apparently vaporized enough of the fuel such that it did not accumulate sufficiently and steady state operation could be achieved. Batch 2 residual fuel showed similar results.

The catalytic reactor was operated for about 16 hours on the residual fuels. No degradation or poisoning of the reactor was observed during this period of testing.

CO EMISSION INDEX

The CO emission index in units of g CO/kg fuel is presented in Figs. 4(a) and (b) as a function of the adiabatic reaction temperature for reference velocities of 10 and 20 m/s. Figure 4(a) presents data for both residual fuels at a reference velocity of 10 m/s, pressures of 3×10^5 and 6×10^5 Pa, and inlet air temperatures from 800 to 960 K. Increasing the adiabatic reaction temperature decreased the CO emissions. Batch 2 residual fuel produced higher CO emissions than batch 1. This was probably caused by the increased dropsize and reduced vaporization of batch 2 as compared to batch 1 residual fuel.

An increase in pressure from 3×10^5 to 6×10^5 Pa decreased the CO emissions. This was probably due to the increased premixing zone temperature caused by preflame reactions and increased gas phase reactions at the higher pressure as previously discussed. As previously shown for combustion efficiency, little effect of inlet air temperature on CO emissions was shown. A comparison of Figs. 4(a) and (b) shows that increasing the reference velocity from 10 to 20 m/s increased the CO emission index for batch 1 fuel at an inlet air temperature of 800 K.

Unburned Hydrocarbons Emission Index

Figures 5(a) and (b) present the unburned hydrocarbons emission index in units of g HC/kg fuel as a function of the adiabatic reaction temperature for reference velocities of 10 and 20 m/s, pressures of 3×10^5 and 6×10^5 Pa, and inlet air temperatures from 800 to 960 K. The results show the same trends as previously shown for the CO emission index. The measured unburned hydrocarbons were extremely low at these relatively high inlet air temperatures. Almost all of the combustion inefficiency, therefore, was caused by the CO emissions.

NO_x Emissions

NO_x emissions, expressed as ppm by volume and corrected to 15 percent excess O₂ are presented in Figs. 6(a) and (b) for both residual fuels at reference velocities of 10 and 20 m/s. Figure 6(a) presents data taken at pressures of 3×10^5 and 6×10^5 Pa, and inlet air temperatures from 800 to 960 K at a reference velocity of 10 m/s. NO_x emissions were found to increase with adiabatic reaction temperature. The NO_x emission standard of 125 ppm by volume at 15 percent O₂ for new, modified, or reconstructed stationary gas turbines is shown for comparison (Ref. 14). It includes an allowance of 50 ppm for fuel-bound nitrogen and is based on a gas turbine thermal efficiency of 25 percent. NO_x emissions were less than the standard for all the test conditions. They ranged from 50 to 110 ppm at 15 percent O₂. No effect of inlet air temperature, pressure, or residual fuel batch is shown. NO_x emissions at a reference velocity of 20 m/s are presented in Fig. 6(b). NO_x emissions at a given adiabatic reaction tempera-

ture were lower for the higher reference velocity, however, combustion efficiency was also reduced.

Figures 7(a) and (b) present the conversion of fuel-bound nitrogen to NO_x as a function of the adiabatic reaction temperature for both residual fuels at reference velocities of 10 and 20 m/s. The conversion was calculated assuming all measured NO_x emissions were due to fuel-bound nitrogen conversion. This was considered to be reasonable because of the relatively low flame temperatures of catalytic combustion. Conversion increased with increasing adiabatic reaction temperature. Conversion levels ranged from 25 to 50 percent. Reference 7 reported conversion levels approaching 100 percent with residual fuel. The fuel used for the study of Ref. 7, however, contained only 0.22 percent nitrogen. Reference 15 reported a strong dependence of fuel nitrogen content on the conversion of fuel-bound nitrogen to NO_x for lean catalytic combustion. Conversion decreased with increasing fuel nitrogen content. At 0.5 percent fuel-bound nitrogen, Ref. 15 reported a conversion to NO_x of 46 percent. Conversion levels for the petroleum - derived residual fuels were lower than those obtained previously for coal-derived fuels (Ref. 3). This was also reported in Ref. 15 for lean catalytic combustion of pyridine doped petroleum-derived ERBS and coal-derived SRCII.

A comparison of Figs. 7(a) and (b) shows the conversion at a reference velocity of 20 m/s was slightly lower at a given adiabatic reaction temperature than at 10 m/s but the combustion efficiency was also lower for the higher reference velocity.

Pressure Drop

Catalytic reactor pressure drop, as a percentage of inlet pressure is presented in Fig. 8 at an inlet temperature of 800 K and a pressure of 3×10^5 Pa for batch 1 residual fuel at reference velocities of 10, 15, and 20 m/s. Pressure drop ranged from about 1.5 percent at a reference velocity of 10 m/s to 3.5 percent at 20 m/s for an adiabatic reaction temperature of 1350 K. This is considered reasonable for an application.

SUMMARY OF RESULTS

A noble metal catalytic reactor was tested with two residual fuels at inlet air temperatures from 700 to 960 K, pressures of 3×10^5 and 6×10^5 Pa, and reference velocities from 10 to 20 m/s. Combustion efficiencies greater than 99.5 percent were obtained from both residual fuels. A more viscous, higher boiling point residual fuel showed poorer performance than a less viscous, lower boiling point residual fuel. An increase of about 70 K in adiabatic reaction temperature was required for the poorer fuel to maintain the same combustion efficiency.

The residual fuels which were used for this study contained from 0.53 to 0.55 percent by weight of fuel-bound nitrogen. For catalytic combustion under the fuel-lean conditions tested, NO_x emissions ranged from 50 to 110 ppm by volume, at 15 percent excess oxygen. Conversion of fuel-bound nitrogen ranged from 25 to 50 percent, a fairly low conversion level.

Successful catalytic combustion of the residual fuels used in this study required inlet air temperatures of at least 800 K to prevent significant

upstream burning in the premixing zone. At inlet air temperatures below 800 K, upstream burning occurred which was probably caused by the accumulation of fuel on the walls of the premixing zone and subsequent ignition of that fuel-air mixture. The increased vaporization at inlet air temperatures of 800 K and above prevented significant deposition of fuel on the duct walls.

The catalytic reactor was operated for about 16 hours on the residual fuels. No degradation or poisoning was observed during this period of testing.

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TABLE I. - DESCRIPTION OF CATALYTIC REACTOR ELEMENTS

Element	Material	Catalyst loading kg/m ³	Substrate manufacturer	Substrate material	Cell density cells/cm ²	Open area, percent
1	Pt/Pd	3.6	Dupont	Mullite	2.6	75
2	Pt/Pd	3.6	Dupont		2.6	75
3	2Pa/1Pt	1.8	Gen. Refractories		10	78
4		1.8			10	78
5		3.6		Cordierite	34	67
6				Mullite	34	67
7			Corning	Cordierite	46.5	63
8			Corning	Cordierite	46.5	63

TABLE II. - DESCRIPTION OF RESIDUAL FUELS

	Batch 1	Batch 2
Elementals, percent by weight		
Carbon	87.16	86.13
Hydrogen	10.65	11.02
Nitrogen	.53	.55
Sulfur	.16	.74
Ash	Not determined	.01
Specific gravity, at 339 K	.899	.864
Flash point, K	434	434
Pour point, K	285	280
Viscosity, CS	101 at 339 K 25 at 367 K	380.7 at 344 K 78.5 at 383 K 38.7 at 433 K
Gross heating value of combustion, J/kg	4.372×10^7	4.365×10^7
Distillation, K percent by volume		
5	530	No distillation to 589 K
10	572	
15	608	

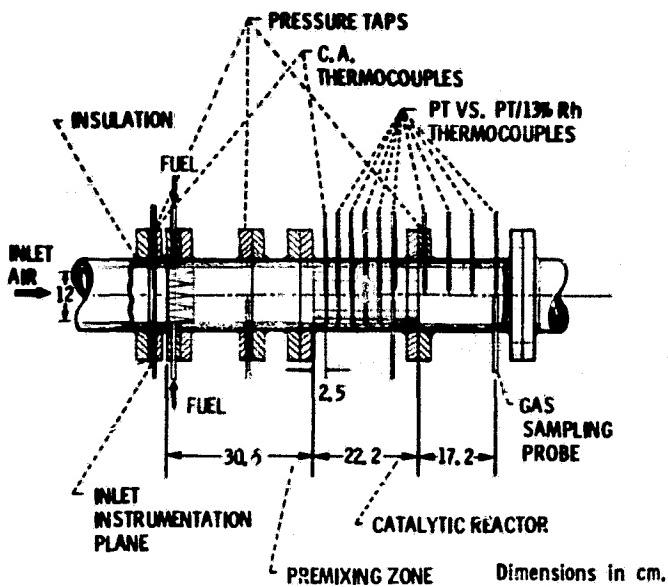
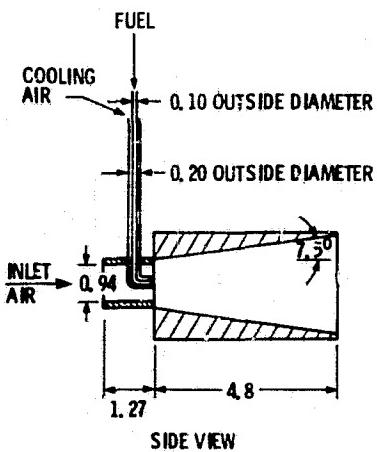
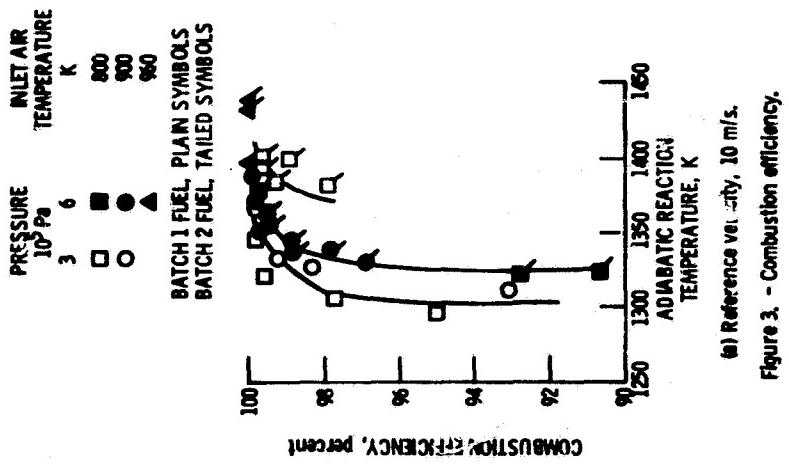


Figure 1. - Test rig schematic.

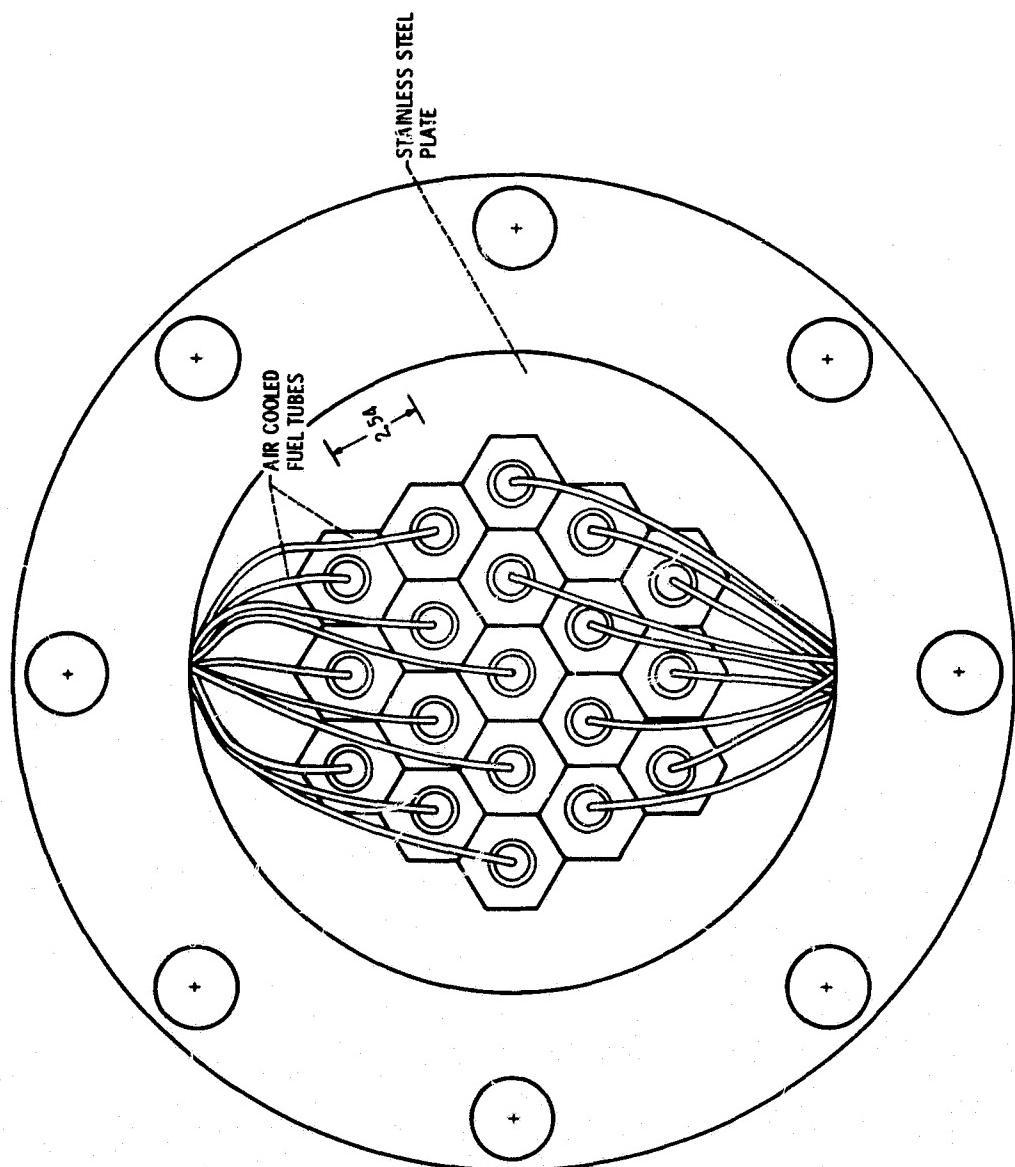


(a) HEX tube fuel injector module (one of 19).
Dimensions in cm.

Figure 2. - Fuel injector.



(a) Reference velocity, 10 m/s.
Figure 3. - Combustion efficiency.



(b) Inlet face. View looking downstream.
Figure 2. - Concluded.

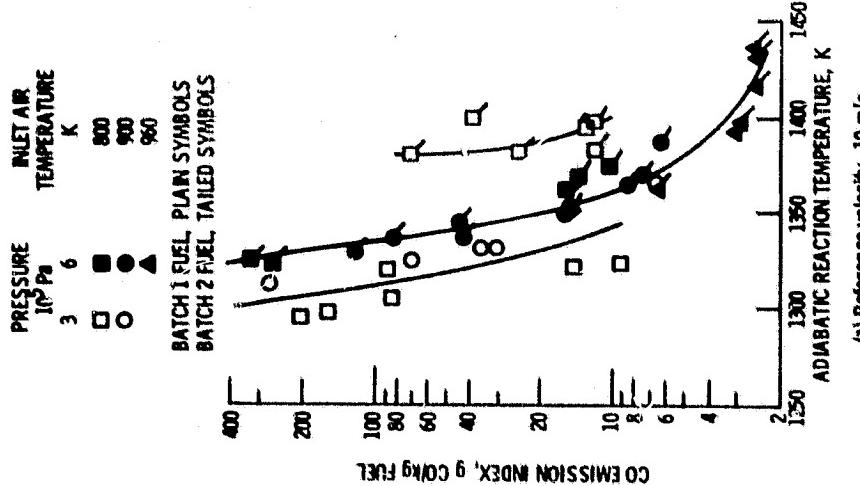


Figure 4 - CO emissions.
(a) Reference velocity, 10 m/s.

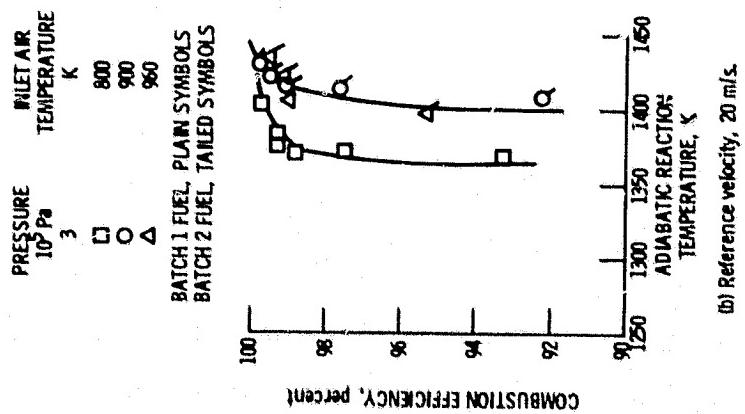


Figure 3 - Concluded.
(b) Reference velocity, 20 m/s.

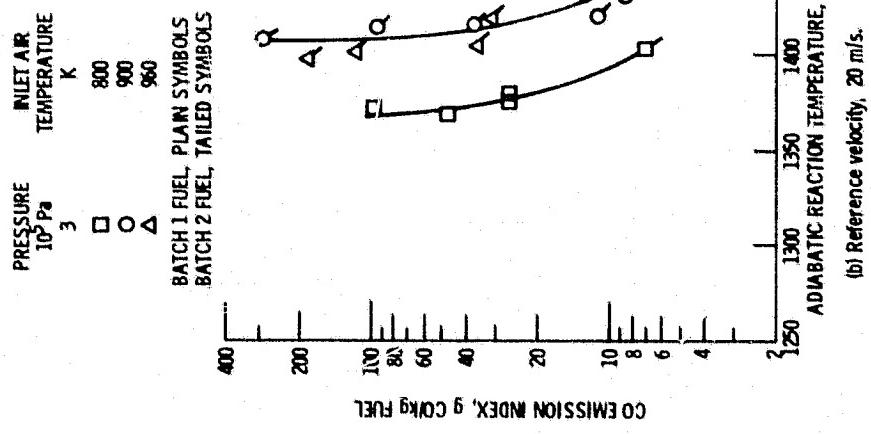


Figure 4. - Concluded.

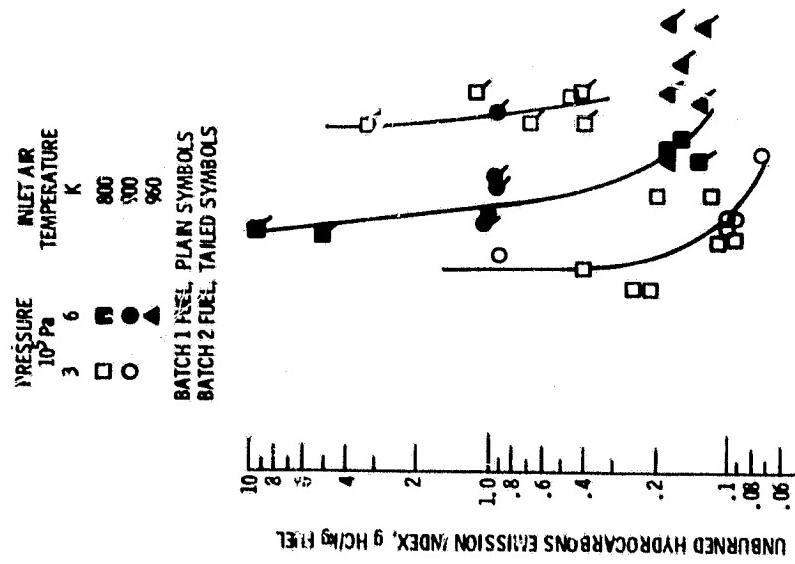
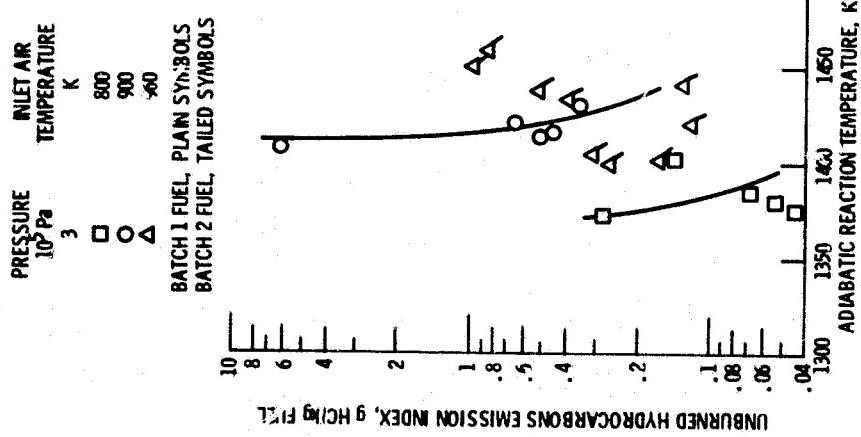
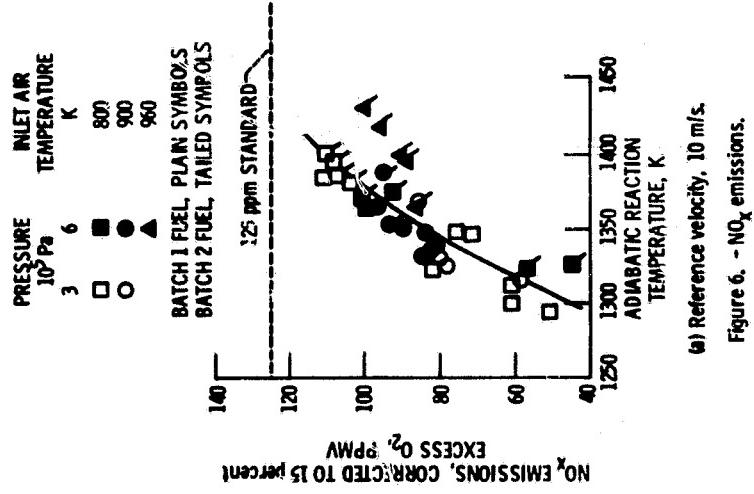


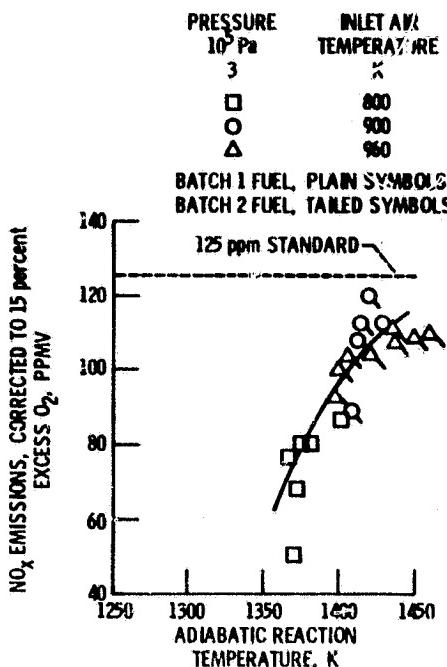
Figure 5. - Unburned hydrocarbon emissions.



(b) Reference velocity, 20 m/s.
Figure 5. - Concluded.

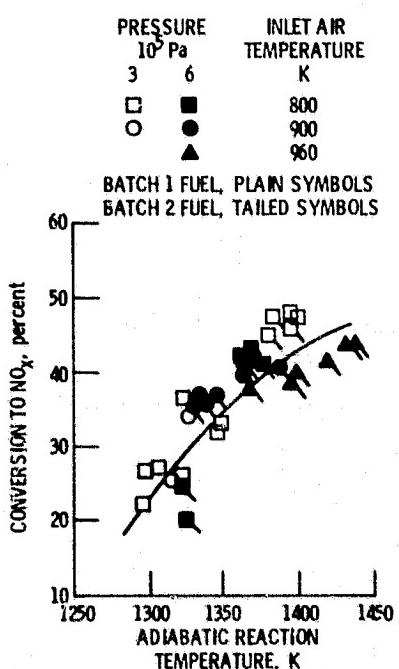


(a) Reference velocity, 10 m/s.
Figure 6. - NO_x emissions.



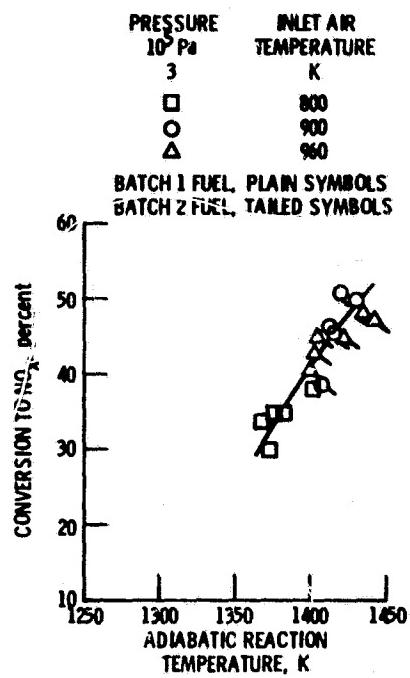
(b) Reference velocity, 20 m/s.

Figure 6. - Concluded.



(a) Reference velocity, 10 m/s.

Figure 7. - Conversion of fuel-bound nitrogen to NO_x.



(b) Reference velocity, 20 m/s.

Figure 7. - Concluded.

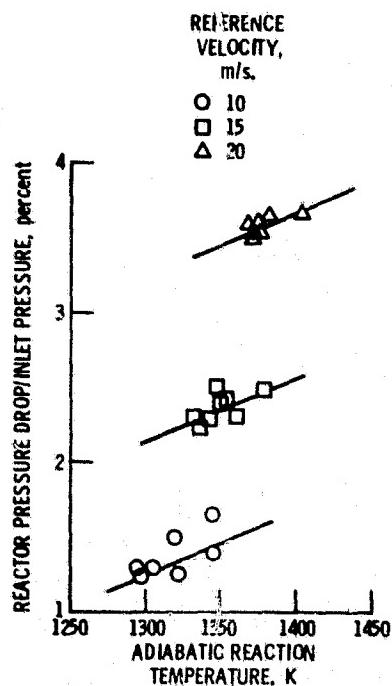


Figure 8. - Effect of reference velocity on pressure drop; inlet air temperature, 800 K; pressure, 3×10^3 Pa; batch 1 residual fuel.